

Final Technical Report ✓

Use of radon and cosmogenic radionuclides as indicators of exchange
between troposphere and stratosphere.

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Award Period: 1 January 1984 - 31 October 1987

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Summary

This research grant covered our participation in the operational phase of NASA's Stratosphere-Troposphere Exchange Project (STEP), a multi-agency airborne science program conducted aboard NASA U-2 and ER-2 High Altitude Research Aircraft. The primary goals of STEP were to investigate the mechanisms of irreversible movement of mass, trace gases and aerosols from the troposphere into the stratosphere, and to explain the observed dryness of the stratosphere. Three flight experiments were conducted to address these questions: two Extratropical Experiments, in 1984 and 1986, and a Tropical Experiment, in 1987. The cosmogenic radionuclides ^7Be and ^{32}P , produced in the stratosphere by cosmic rays, and ^{222}Rn (radon), emitted from continental soils, were well-suited as tracers of intra-stratospheric air mass movements, and to follow episodes of troposphere-to-stratosphere exchange. Measurements of ^7Be and ^{32}P were made in all three STEP Experiments. Measurements of radon were made in the Tropical Experiment only. Our equipment worked well, and produced a valuable data set in support of the STEP objectives, as indicated by the "quick-look" results outlined in the report.

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Summary

- 1.0 Introduction and overview
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1.0 Introduction and overview

While there is general agreement that most air entering the stratosphere does so in the tropics, and that the cold temperatures characteristic of the tropical tropopause are responsible for the observed dryness of the stratosphere, there has been a continuing debate about the actual mechanism of the transport (e.g., slow regional upwelling versus fast localized convection) (e.g., Brewer, 1949; Dobson, 1956; Kritz, 1975; Newell and Gould-Stewart, 1981; Danielsen, 1982 and Holton, 1984). These questions were directly addressed by the STEP program, whose primary goals were to investigate the mechanisms of irreversible transport of mass, trace gases and aerosols from troposphere to stratosphere (at midlatitudes as well as in the tropics) and to explain the observed dryness of the stratosphere.

To achieve these ends a team of 14 principal investigators was assembled under the scientific leadership of Dr. Edwin Danielsen. Two types of flight experiments---Extratropical and Tropical---were conducted. Two Extratropical Experiments were conducted from Moffett Field, CA, in 1984 and 1986, while a Tropical Experiment was conducted from Darwin, Australia in 1987. Atmospheric properties and species measured during the course of the experiments included aerosol size spectra, condensation nuclei, water vapor and total water, ^7Be , ^{32}P and ^{222}Rn (radon),

ozone, air motions (temperature, pressure and winds), reactive nitrogen (NO_y), carbon monoxide (CO), infrared radiation and total radiance, and local vertical temperature profiles.

Although two precursor experiments had been conducted from Panama in 1977 and 1980 (Margozi, 1983) aboard a U-2 aircraft with a much smaller instrument complement, the STEP experiments represented a quantum jump forward in terms of the larger suite of atmospheric properties measured, and the scope and intensity of the flight program. Thus the principal activities of the Principal Investigator Team (and for our group as well) during the operational phase of STEP centered on discussions of how best to conduct the flight experiments, instrument development, the actual conducting of the three flight experiments, and preliminary interpretation of the results from the 1984 Extratropical Experiment. These preliminary interpretations, in the approximate state of development they were in at the close of the operational phase of STEP (and this research grant) are outlined below. These preliminary analyses were extended and expanded in the (open-ended) STEP data analysis phase. Two special journal issues of peer-reviewed papers have been published thus far in the STEP data analysis phase, in 1990 and 1993.

2.0 Role of the natural atmospheric radionuclides in the STEP Experiments

2.1 ^7Be and ^{32}P . The cosmogenic radionuclide ^7Be is produced in the atmosphere (primarily the stratosphere) by interactions between cosmic rays and oxygen and nitrogen. The local production rate is a function of the local atmospheric density, the local cosmic ray flux and energy spectrum, and the cross sections for the various relevant interactions. These have been determined as a function of geomagnetic latitude and altitude (c.f. Bhandari, et al., 1966, Lal and Peters, 1967) as shown in Figure 1.

Upon formation, the newly formed atoms of ^7Be are quickly and irreversibly scavenged by sub-micron sized particles of the ambient atmospheric aerosol, and so are subject to removal and transport by the same processes affecting the aerosol. This is a minor consideration in the lower stratosphere, where sub-micron aerosol sedimentation velocities are small and aerosol residence times many times greater than ^7Be 's 53 day radioactive half-life. However this situation is reversed in the troposphere, where the importance of precipitation scavenging and the consequent short (~1 week) aerosol residence times result in the removal of most tropospheric ^7Be by

Radionuclide concentrations are frequently expressed as activities, as expressed by the formula $A=N\lambda$. Here A is the activity, [radionuclide disintegrations per unit time, e.g. dpm], N the number (or concentration) of atoms of the radio-nuclide, and λ the radioactive decay constant for that radionuclide. Thus a ^7Be concentration of 10^6 atoms/scm is equivalent to an activity of 9 dpm/scm. Atmospheric radionuclide activities are sometimes expressed in pico-Curies (pCi) where 1 pCi = 2.2 dpm, or Bequerels (Bq) where 1 Bq = 60 dpm.

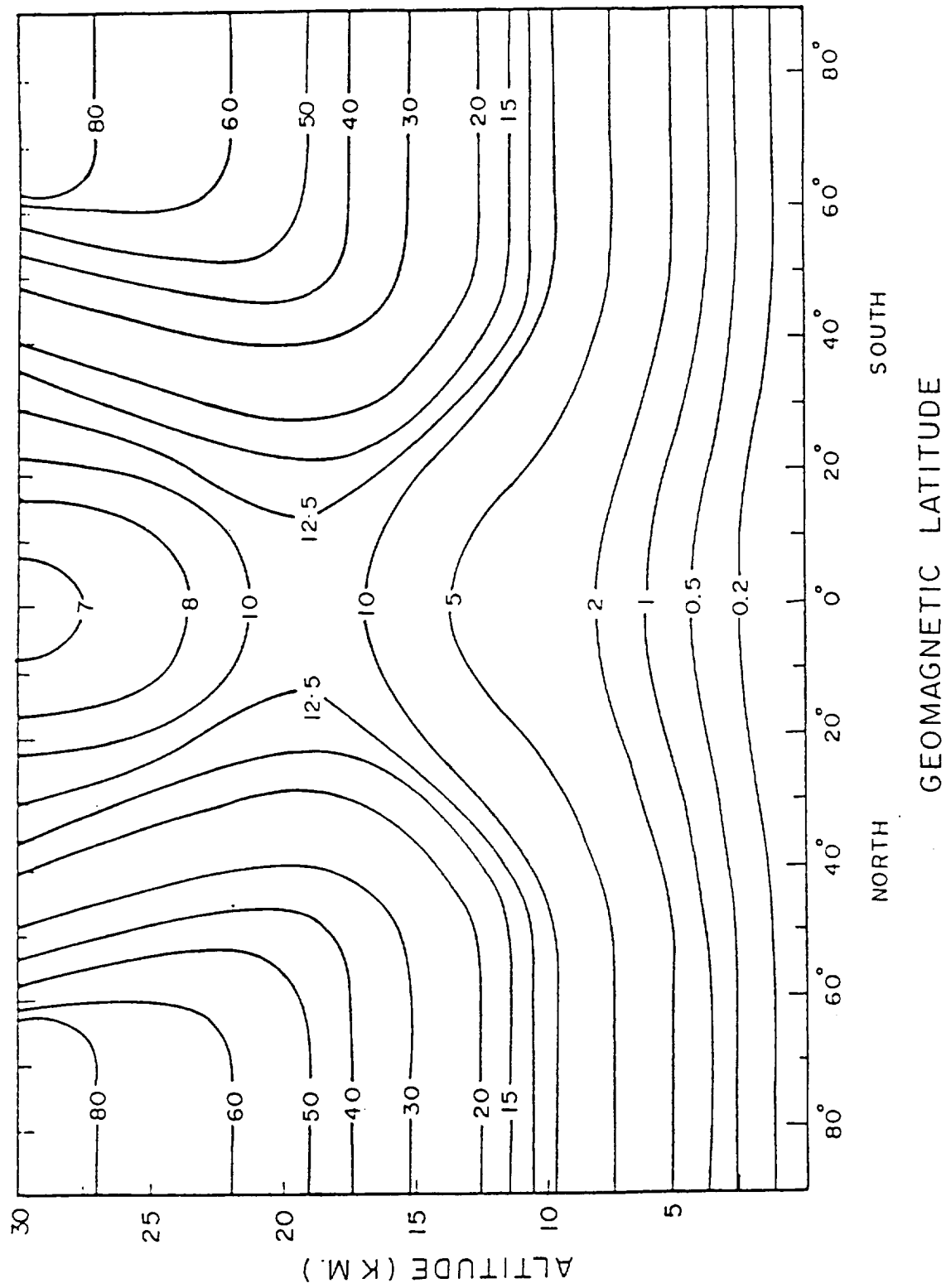


Figure 1: ^7Be production as a function of altitude and geomagnetic latitude. Units are disintegrations per minute per standard cubic meter (dpm/scm). After Bhandari et al., 1966

scavenging rather than by radioactive decay. Thus in the mid-latitude and tropical lower stratosphere ^7Be concentrations (or activities; see note below) tend to approach the local equilibrium value, on the order of 10 or 20 dpm/scm, as borne out by our STEP ^7Be measurements as well as those compiled by Bhandari et al. [1966] and Krey [1967]. As a result of rapid scavenging by precipitation, ^7Be activities in the upper troposphere are typically ≤ 1 dpm/scm, a fraction of the local equilibrium value. Because of these considerations, the convenient length of its radioactive half-life, and the absence of chemical sources or sinks, ^7Be is an excellent tracer of stratospheric air in the troposphere and of large-scale intra-stratospheric motions (c.f. Bhandari et al., 1966). Conversely, the *absence* of ^7Be in the stratosphere is indicative of a recent entrainment of tropospheric air.

Most of these considerations also apply to ^{32}P , which is produced by interactions between cosmic rays and argon and has a 14 day half life. The production function of ^{32}P is quite similar to that of ^7Be (see Figure 1) the major difference being that at any given point the local production rate of ^{32}P is only about 1/110 that of ^7Be . (Actually this ratio varies by about 10% over the extent of the stratosphere. While not relevant to the present discussion it is taken into account in our calculations.)

If the atmosphere were motionless, the local concentration of each of the cosmogenic radionuclides would reflect the equilibrium between its local rate of production and its rate of radioactive decay. If this concentration is expressed as an activity, as discussed earlier, the equilibrium activity of a given radionuclide is identical with its rate of production. Thus in a motionless atmosphere the equilibrium $^7\text{Be}/^{32}\text{P}$ activity ratio would be ~ 110 throughout the troposphere and lower stratosphere. However in the real atmosphere two processes occur which tend to prevent this equilibrium from being attained. First, both ^7Be and ^{32}P are removed by sedimentation and scavenging of the particles to which they are attached. Second, because the real atmosphere is in motion, the radionuclide concentrations observed at a given location are sometimes more characteristic of the region from which that air was recently transported than they are of the region of observation—which of course forms the basis for their use as tracers. However if one considers the *ratio* of a suitable radionuclide pair, such as $^7\text{Be}/^{32}\text{P}$ another, more dramatic effect can occur.

To illustrate this point we consider a motionless atmosphere with no cosmic ray flux. If at $t=0$ the flux is turned on, the local concentration, N , of the various radionuclides will start to build up and to approach their equilibrium values, N_∞ , as described by the equation:

$$N = N_\infty (1 - e^{-\lambda t}),$$

or, in terms of activity, (recall $A = N\lambda$):

$$A = A_{\infty} (1 - e^{-\lambda t}).$$

Thus after a period of time equal to 3 radioactive half-lives the local activity of a given radionuclide would have attained approximately 88% of its equilibrium value. For ^{32}P , with a half-life of 14 days, this would occur in 42 days; for ^7Be , with a 53 day half life, 159 days would be required. Thus for times greater than, say, 180 days both radionuclides would be close to equilibrium and the $^7\text{Be}/^{32}\text{P}$ activity ratio would be quite close to their production ratio; i.e. ~ 110 .

If, after equilibrium had been reached, the cosmic ray flux were to fall to zero the local activity would *decrease* with time, as described by the equation:

$$A = A_{\infty} (e^{-\lambda t}).$$

Thus 28 days after this hypothetical cosmic ray cut off the ^{32}P activity would have dropped by $\sim 75\%$, to $\sim 25\%$ of its equilibrium value. In this interval ^7Be , due to its longer half life, would have diminished by only $\sim 30\%$, i.e. to about 70% of its equilibrium value. However at this point the $^7\text{Be}/^{32}\text{P}$ activity *ratio* would have risen from its equilibrium ratio of ~ 110 to an intermediate value of ~ 300 !

An analog to this process in the real atmosphere is the movement of a parcel of equilibrated air from a higher latitude, where cosmic ray fluxes and hence radionuclide production rates are high (see Figure 1) to a lower latitude, where production rates are lower, which would result in an upward excursion of the $^7\text{Be}/^{32}\text{P}$ activity ratio. Similarly, the movement of equilibrated air from lower to higher latitudes would result in a downward excursion of the $^7\text{Be}/^{32}\text{P}$ activity ratio to significantly below 110. Several such excursions of the $^7\text{Be}/^{32}\text{P}$ activity ratio were in fact observed during the STEP experiments. While interpretation of these excursions was not possible during this, operational, phase of the STEP program, they are of particular interest as they may be indicative of rapid intra-stratospheric meridional movements.

2.2 Radon. Radon (^{222}Rn) enters the atmosphere from the earth's crust, where it is produced by the radioactive decay of trace quantities of its parent, ^{226}Ra . (Radon emission rates from the sea are less than 1% of the land values.) Because radon is a noble gas and relatively insoluble in water its removal from the atmosphere by precipitation is negligible, so that its only significant atmospheric sink is its own radioactive decay, which occurs with a half-life of 3.8 days. For these reasons atmospheric radon measurements have been used to derive coefficients of vertical mixing (e.g. Liu et al., 1984), as well as an indicator of the transport of continental air over the oceans (e.g. Prospero and Carlson, 1970).

The highest concentrations of atmospheric radon occur in the continental boundary layer, where values in the range of 100-300 pCi/scm are frequently observed (Gold et al, 1964; Lambert et al. 1982). Radon concentrations in the free troposphere are typically an order of magnitude smaller, decreasing, in the mean, with altitude to values on the order of 1 pCi/scm near the tropopause (Moore et al., 1973; Liu et al., 1984). Radon activities in stratosphere are essentially zero (Moore et al., 1973) except, as shown by our STEP results, during and shortly after episodes of troposphere-to-stratosphere transport. Thus radon was ideally suited as a tracer for tropospheric air in the STEP experiments. In particular, the observation of stratospheric air with high radon and low total water contents would be unambiguous proof of the recent tropospheric origin of that air, and of the presence of an effective dehydration process acting in conjunction with or shortly after the movement into the stratosphere. Several such instances of high radon and low total water were observed during the STEP Tropical Experiment, as discussed in Section 4.

3.0 The 1984 and 1986 Extratropical Experiments

These experiments, comprising four flights each, were conducted from Moffett Field in April and May of 1984 and 1986, and focused on transports and exchanges associated with cyclogenesis in the vicinity of mid-latitude upper tropospheric jets. The in situ observations made aboard the NASA U-2 in the 1984 Experiment included meteorological state variables (pressure, temperature and horizontal winds) and trace constituent concentrations (ozone, water vapor, condensation nuclei, ^7Be and ^{32}P).

At the time of the end of this project (i.e. the end of the operational phase of STEP) a preliminary assessment of the ^7Be measurements made in the 1984 experiment had been made to attempt to identify entrainments of upper tropospheric air into the lowest few kilometers of the mid-latitude stratosphere (c.f. Danielsen, 1968). That analysis is outlined below.

Figure 2 shows the U-2 flight path relative to the jet core and tropopause fold structure based on the analysis of Danielsen from the in situ measurements made above the jet core on the April 20, 1984 flight. On this date the NASA U-2, CV-990, and Electra aircraft, under the direction of E. F. Danielsen, flew simultaneous, coordinated flight paths at various levels in the jet and fold structure (e.g., Hipskind et al., 1987).

The ^7Be activities of the samples collected at 12.5 km altitude (41,000 feet) at $\sim 35^\circ\text{N}$, just above the jet core, a region of strong vertical and horizontal wind shear, were similar to those which would result from an ongoing mixing process involving tropospheric air from the anti-cyclonic (equatorial) side of the jet and stratospheric air from the cyclonic (polar) side of the jet. An

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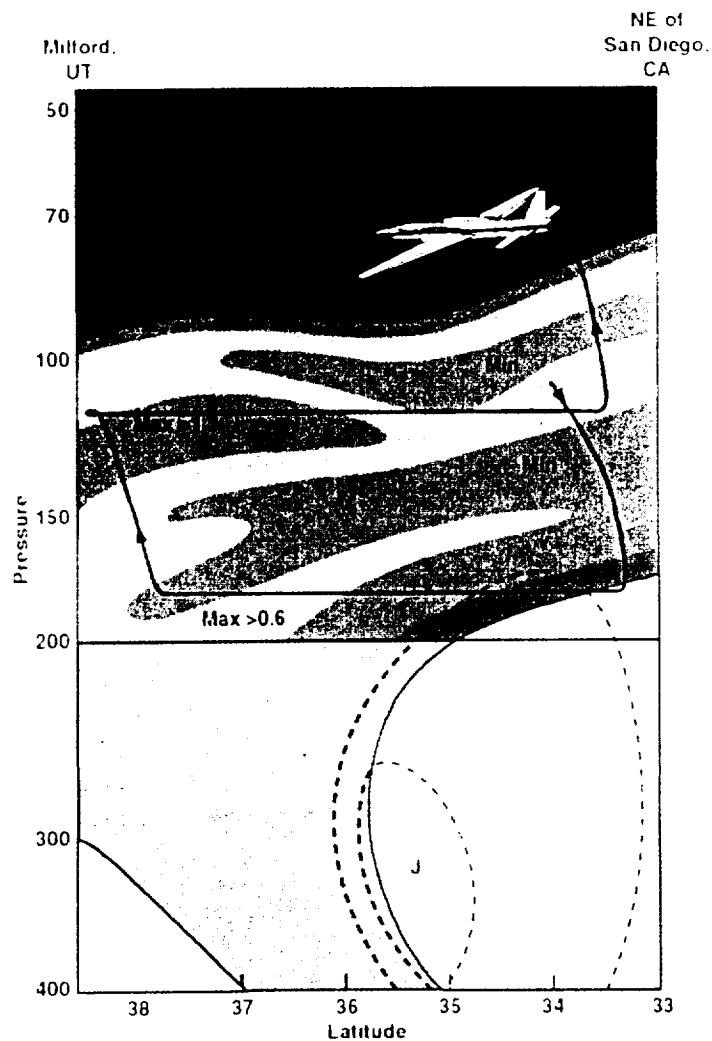


Figure 2. 1984 Extratropical Experiment. U-2 flight path relative to the jet core and tropopause fold structure on April 20, 1984, as analyzed by E. F. Danielsen.

analysis based on simple linear mixing of ^7Be indicated a ~45% tropospheric contribution; parallel analyses based on simultaneous observations of ozone, water vapor and potential vorticity yielded tropospheric coefficients of 52%, 42% and 42%, respectively. Similar close correlations were observed in the 12.5 km leg of the May 6 flight, which had a similar flight pattern and was flown in approximately the same position relative to a similar jet/fold structure.

These results are indicative of an entrainment of upper tropospheric air into the lower stratosphere, a process believed to be important (if not dominant) in maintaining the observed vertical gradients of many trace constituents in the lower few kilometers of the mid-latitude stratosphere. For example, the vertical profiles of water vapor observed by Kley et al. (1979) at mid-latitudes do not show a discontinuous change in slope or value at the tropopause, but rather continue to decrease with altitude, reaching a minimum value (the hygropause) several kilometers above the tropopause. In this region between the hygropause and the tropopause (the transition zone) stratospheric properties of the air appear to be diluted by increasing proportions of tropospheric air at the tropopause is approached, a situation which is with the preliminary analysis, outlined above, of our ^7Be results on the April 20 and May 6 flights.

4.0 The 1987 Tropical Experiment

As mentioned earlier, there has been a continuing debate over the actual mechanism of troposphere-to-stratosphere transport in the tropics, and also whether the dehydration process responsible for the observed dryness of the stratosphere occurs in conjunction with this movement. These questions were directly addressed by the STEP Tropical Experiment, which focused on the troposphere-to-stratosphere movement of air in the tropics, with particular emphasis on exchange and dehydration processes associated with tropical cirrus and cumulonimbus clouds.

Darwin, Australia, in a January-February timeframe was chosen as the site for the STEP Tropical Experiment because of the extremely cold upper tropospheric and lower stratospheric temperatures, and intense thunderstorms, characteristic of the region and season. Eleven flights were flown from Darwin, sampling the upper-level outflows from mesoscale convective and cyclonic systems, penetrating the cirrus anvils of several cyclonic storms, and characterizing background conditions in the absence of nearby large convective systems.

Preliminary analyses of the in situ radon and other trace constituent measurements made in and downwind of the cirrus shields of three large tropical cyclones (Flights 5, 6 and 11) and downwind of the cirrus anvil of a large cumulonimbus cloud cluster showed what appeared to be

several clear instances of elevated radon activities occurring simultaneously with low total water mixing ratios. These observations appear to be clear evidence of an effective dehydration process, capable of reducing total water mixing ratios to less than 2.5 ppmv occurring in conjunction with troposphere-to-stratosphere exchange.

Samples for ^7Be and ^{32}P measurements were also collected during the Tropical Experiment. These samples have been analyzed, but as of the end of the operational phase of STEP (and of this research grant) had not been interpreted.

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